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Linköping University Post Print

N.B.: When citing this work, cite the original article.

Original Publication:

Alessio Perrone, M D'Elia, M. Di Giulio, G. Maruccio, A. Cola, N. Stankova, D. Kovacheva and Esteban Broitman, Non-conventional Cu thin films deposited on Y substrate by sputtering, 2014, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, (), 752, 27-32.

<http://dx.doi.org/10.1016/j.nima.2014.03.032>

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Postprint available at: Linköping University Electronic Press

<http://urn.kb.se/resolve?urn=urn:nbn:se:liu:diva-106598>

Non conventional photocathodes based on Cu thin films deposited on Y substrate by sputtering

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ABSTRACT

Copper (Cu) thin films were deposited on yttrium (Y) substrate by sputtering. During the deposition, a small central area of the Y substrate was shielded to avoid the film deposition and was successively used to study its photoemissive properties. This configuration has two advantages: the cathode presents (i) the quantum efficiency and the work function of Y and (ii) high electrical compatibility when inserted into the conventional radio-frequency gun built with Cu bulk. The photocathode was investigated by scanning electron microscopy to determine surface morphology. X-ray diffraction and atomic force microscopy studies were performed to compare the structure and surface properties of the deposited film. The measured electrical resistivity value of the Cu film was similar to that of high purity Cu bulk. Film to substrate adhesion was also evaluated using the Daimler-Benz Rockwell-C adhesion test method. Finally, the photoelectron performance in terms of quantum efficiency was obtained in a high vacuum photodiode cell before and after laser cleaning procedures. A comparison with the results obtained with a twin sample prepared by pulsed laser deposition is presented and discussed.

Keywords: sputtering, thin films, electrical resistivity, photocathodes, quantum efficiency, adhesion strength.

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Introduction

Metal-based photocathodes are being used in the development of high brightness electron beams [1-3]. Since metallic photo-cathodes display multiple advantages (long lifetime, short response time and robustness), research has been focused on developing new types and low cost devices. Recent research has also showed that metallic thin films exhibit good photo-emissive properties [4-8]. However, despite their large applications there have been only few methods reliable and available to generate metallic thin films with fine control over their photo-emissive performances.

Recently, some studies have shown that laser ablation and deposition in high vacuum of Mg [9] and Y [10] thin films is reliable and easily applicable, avoiding additional steps as annealing, necessary for other thin film deposition techniques, that could increase the surface contamination. Unfortunately the metallic photocathodes also suffer of surface contamination when exposed to atmosphere, even if much less than semiconductor photocathodes [11].

Therefore, laser cleaning treatments are mandatory for the rejuvenation of the cathode surface but, in the case of thin film photocathodes, these treatments can lead to the delamination process or the consumption and thinning of the film. In order to overcome such drawbacks, a new configuration of metallic photocathode was proposed [12]. In that configuration a disk of Y bulk was covered by a film of Cu deposited by pulsed laser deposition technique (PLD), while masking the area of the photo-emitting spot, i.e. the central part of the Y disk. The Cu PLD films presented a droplet density higher than $10^4/\text{mm}^2$, characteristics of PLD metallic films. Accordingly, this new configuration pursues the aim of obtaining a metallic photocathode with the superior quantum efficiency (QE) of Y and the excellent electrical resistivity of Cu, reported in table I.

Herein we report for the first time the deposition of Cu film on Y disk by sputtering technique, with the aim to reduce the droplet density on the film surface, improving the electrical surface resistivity of the thin film. We also investigated its photoemission properties in terms of quantum efficiency (QE) at different experimental conditions.

Experimental procedures

Cu films have been deposited on Y disks by RF-diode sputtering, starting from a pure (99.99%) Cu 13 cm in diameter target. The ultimate vacuum of the cryo-pumped vacuum system was 2×10^{-5} Pa. The process gas was ultrahigh purity argon (99.9998%) at a pressure of 0.67 Pa that was introduced in the deposition chamber through a mass flowmeter rated at 50 sccm. The target-substrate distance was 5.5 cm and the RF power was 500 W, resulting in a deposition rate of about 43 nm/min. The Y substrate was masked in the center by a metal cone of 6 mm in diameter and 5 mm in height, as shown in Fig. 1.

In the same deposition run, besides the Y disk other small glass sheets were fully covered with the same Cu film for further characterization and thickness measurements. All the substrates were placed on a water-cooled holder and kept at room temperature.

The deposition process was continuously monitored by means of an in-situ Maxtek quartz microbalance previously calibrated to read the real thickness value.

Scanning electron microscopy (SEM, model JEOL-JSM-6480LV) was used for the analysis of the film morphology, whereas X-ray diffraction (XRD, Bruker AXS D8 Advance X-Ray diffractometer using Cu $K\alpha$ line) was utilised for the characterisation of the structure and crystal orientation of the deposited film. X-ray diffraction scans were taken in the large angle (20° - 120°) $\Theta/2\Theta$ mode with a constant step 0.02° and counting time of 35 s/step. Phase identification was performed with the *DiffraPlus* EVA using ICDD-PDF2 Database. Topographic analysis of the sample surface was performed using a commercial atomic force microscope (AFM, Bioscope 2, Veeco Instruments Inc. Santa Barbara, CA, USA), which was operated in contact mode at room temperature in ambient air using V-shaped silicon nitride cantilevers with a nominal spring constant of 0.01 N/m (MLCT-AUNM, Veeco).

The compositional map of the surface was deduced by means of energy dispersive X-ray (EDX, IXRF

Systems EDS Sirius SD) spectrometer coupled with the SEM. The thickness of the Cu thin film deposited on a quartz substrate in the above experimental conditions was measured in different points by a profilometer (Tencor Alphastep). Electrical resistivity measurements (Ecopia 3100 system) were carried out on this sample in the Van der Paw configuration. For this purpose gold tips were placed on the four corners of the squared quartz sample. The measurements were performed in air and at room temperature. Finally, adhesion of the film to the substrate has been evaluated by the Daimler-Benz Rockwell-C adhesion test method [13]. Indentation was done with a standard Rockwell hardness tester fitted with a Rockwell C-type diamond cone indenter with an applied load of 150 N. The adhesion result is obtained by using an optical microscope and classifying the adhesion failures as HF1 to HF6 according to the level of cracking and coating delamination around the indent [13,14]. The HF1 result corresponds to the best adhesion, with very little or no cracks, while HF6 has the worst adhesion result, with big delaminated areas and long cracks. After the characterization, the quantum efficiency of the sample was tested by a home-made photodiode cell.

Results and discussion

Parametric studies were carried out to optimize the deposition process of Cu film on Y substrate. All the deposited films grown at different experimental conditions were adherent to their substrate. Different samples were prepared but only one was sampled out to be analyzed. The film prepared with the optimized experimental parameters was studied with an array of analyses and tested as photocathode by a photodiode cell.

Figure 2 shows the photo of the cathode prepared by sputtering technique shielding the central part of the Y disk with a metal cone of 6 mm in diameter and 5 mm in height. The thickness of the Cu film was measured by a profilometer in different points. The measured average value was around 530 ± 15 nm.

The relief of the Y substrate surface was not completely covered by the Cu film as shown in Fig. 3a. However, the surface morphology of the film is free of droplets, in opposition to the Cu film deposited by PLD, which was covered by round-shape droplets as shown in Fig. 3b.

Two-dimensional maps of Y, Cu and O distributions on the sample surface were obtained by EDX. The SEM-EDX maps are reported in Fig. 4. They were obtained with relative high electron energy (20 keV) because the thickness of the Cu film was large enough to avoid the contribution of the X-rays coming from the Y substrate. The top part of the image relative to EDX map of Y (see Fig. 4) appears black because no X-rays coming from the substrate have been detected. On the contrary, the bottom area coloured in blue shows the uncoated Y substrate. The map of Cu does not seem to present evident voids, which means that the film has been deposited quite uniformly. The limited sensitivity of EDX to oxygen and low level of oxidation are witnessed by the low intensity of red color. The oxygen map shows a higher concentration of this element in the substrate uncoated area. The reason for this effect is probably due to the higher chemical reactivity of Y respect to Cu with the oxygen-containing molecules of the atmosphere.

X-ray diffraction and atomic force microscopy investigations were carried out to deduce the structure and surface morphology of the deposited film.

Figure 5a shows Θ - 2Θ XRD pattern of the Cu film deposited by sputtering on polycrystalline polished Y substrate. The diffraction peaks at 2Θ values 43.3° and 50.5° were identified as (111) and (200) planes of Cu cubic structure, respectively, on the base of ICDD-PDF2 01-071-3761 card. The peaks corresponding to the Y polycrystalline substrate are evident. Highly preferred orientation along (111) direction can be seen, whereas the other diffraction peak being negligible. The mean size of the coherently scattering domains of Cu is 37 nm. Mean crystallite size was determined with the Topas-4.2 software package using the fundamental parameters for peak shape description including appropriate corrections for the instrumental broadening and diffractometer geometry. Using the Debye-Scherrer formula [15], the mean size value of the crystallites oriented along (111) direction was calculated to be smaller at about 31 nm

because the instrumental broadening was not taken into account. Whereas, the mean size of the crystallites grown by PLD along (111) direction was calculated by using the same formula to be at about 25 nm. The XRD pattern of the sample prepared by PLD is shown in Fig. 5b. The higher crystallites dimensions as well as the considerably high intensity and the sharpness of the (111) XRD peak are indicative of better crystal structure of the Cu film obtained by sputtering process.

AFM analyses were also performed to evaluate the film morphology and rms roughness. To assess the homogeneity of the film, different area of the sample were probed. Figure 6a shows a typical AFM image. After Cu deposition, the substrate morphology significantly changes: lapping grooves become less evident and the sample exhibits a more granular aspect, with a typical grain size around 100 nm, which taking into account tip convolution effects can be reduced to about 60-70 nm. These features are reproducible on the whole Cu film, independently on the imaged area. The histogram reported in Figure 6b provides an estimation of the rms roughness value, which is around 40 nm. The size of the grains estimated by AFM measurements is thus reasonably compatible with estimations from XRD data.

The adhesion test (Fig. 7) revealed no visible delamination or cracks around the indentation crater, which is typical of the optimal adhesion strength quality HF1. The excellent adhesion of the Cu film on the Y substrate can be explained by two factors: a) the films have been deposited by sputtering, which is a technique that promotes an increased adhesion of films on many different kinds of substrates [16], and b) it has been previously shown that Cu films deposited at room temperature over Y form an amorphous $\text{Cu}_x\text{Y}_{1-x}$ phase [17], and such inter-diffusion could also contribute to an increased adhesion.

The sheet resistance of the Cu film was around $3.1 \times 10^{-1} \Omega/\text{sq}$ and the corresponding resistivity was $(1.6 \pm 0.1) \times 10^{-6} \Omega \text{ cm}$. The good homogeneity of the film was confirmed by several measurements performed on different area of the deposited film. The average value of the resistivity is similar, in the experimental error, to that of high purity Cu bulk ($1.69 \times 10^{-6} \Omega \text{ cm}$) but much lower than the resistivity value of the Cu film deposited by PLD [12].

A careful laser cleaning process of the sample was carried out in order to reach the maximum QE possible. During such process, carried out by 900 laser pulses, the fluence was fixed below the ablation threshold in order to preserve the photocathode surface. Before the laser cleaning process, the QE of the sample, under 266 nm laser irradiation, was measured to be 1.2×10^{-5} . However by increasing the number of pulses (with an energy density of 0.3 J/cm^2) used for the cleaning process, the QE was enhanced up to a maximum of 6.4×10^{-5} , as we can see in Fig 8. Amounts of laser pulses larger than 900 worsened the photoemission performance of the sample; in fact, as can be observed in Fig. 8, the QE decreased down to 3.0×10^{-5} when the laser cleaning was performed with 6000 laser pulses. This fact can be attributed to a deterioration of the substrate surface morphology by an excessive number of laser pulses. Hence the adequate selection of the energy density and number of laser pulses will allow us to reach the maximum QE of the photocathode. The relatively low QE of the cleaned sample, compared to the value of bulk pure Y [18], can be attributed to the morphology of the Y substrate surface; the lack of a perfectly smooth photocathode surface could provoke a decrease on the photoemission performance of the cathode. Therefore, by improving the substrate morphology, the actual QE of bulk Y could be reached obtaining a satisfactory photoemission performance. On the other hand, under the vacuum conditions of the photodiode cell ($P_{\text{total}} = 4 \times 10^{-6} \text{ Pa}$, $P_{\text{H}_2\text{O}} = 3 \times 10^{-7} \text{ Pa}$, $P_{\text{O}_2} = 7 \times 10^{-8} \text{ Pa}$, $P_{\text{H}_2} = 6 \times 10^{-8} \text{ Pa}$), the QE of the photocathode decreases considerably during the first 10 minutes after the laser cleaning process, showing afterwards a much slighter slope (Fig. 9). This behavior can be attributed to the quick oxidation process of the photocathode surface that, once a thin oxide layer is grown on the yttrium surface, tends to slow down with the time. Accordingly, the use of such photocathodes in an improved vacuum environment, with lower total pressure and partial pressures of reactive molecules such as O_2 , H_2 and H_2O , will lead to a longer lifetime of the photocathode with an adequate QE.

Conclusion

In this paper we use sputtering technique to deposit Cu film on Y substrate forming a new configuration of photocathode. The Cu film was free of droplet and very adherent to its substrate. EDX maps confirm

the uniformity of the deposited film and its low chemical reactivity with the molecules of the air. The electrical resistivity value of the film is comparable to that of Cu bulk indicating that the photocathode is appropriate to be inserted in an rf cavity. We also show that the laser cleaning treatment becomes crucial for improvement the QE but it is important to prevent the deterioration of surface morphology. Finally, the contamination process of the Y surface was invoked to justify the decrease of QE in the time.

In conclusion, the morphological and electrical properties of Cu films deposited by sputtering are more performing than those prepared by PLD technique in the ns regime.

Acknowledgments

This work was supported by the Istituto Nazionale di Fisica Nucleare (INFN). Esteban Broitman acknowledges the Swedish Government Strategic Research Area in Materials Science on Functional Materials at Linköping University (Faculty Grant SFO-Mat-LiU # 2009-00971).

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252 [18] J. Fisher, T. Srinivasan-Rao, T. Tsang, Workshop on Short Pulse High Current Cathodes, Bendor
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258 Table I. Physical properties of Cu and Y bulk.

	Cu	Y
Quantum efficiency	1.4×10^{-4} at 266 nm	5×10^{-4} at 266 nm
Work function	4.5 eV	3.0 eV
Electrical resistivity	$1.72 \times 10^{-8} \Omega \text{ m}$	$59.6 \times 10^{-8} \Omega \text{ m}$

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Figure captions

Fig. 1. Schematic of the standard sputtering system.

Fig. 2. Photo of the sample obtained by the deposition of Cu thin film on Y disk substrate. The central part of the cathode is uncovered in order to induce the photoemission from the Y bulk.

Fig. 3. SEM micrograph of the Cu thin film: a) deposited by sputtering; b) deposited by PLD.

Fig. 4. SEM image and EDX maps of the cathode obtained with electron energy of 20 keV.

Fig. 5. θ -2 θ XRD pattern of the final deposited Cu film on the Y substrate: a) deposited by sputtering; b) deposited by PLD.

Fig. 6. a) AFM image; b) histogram of rms roughness values.

Fig. 7. Indentation crater left by the adhesion test.

Fig. 8. Collected charge of the sample recorded as a function of the laser cleaning shots. The error bars indicate the maximum errors.

Fig. 9. Normalized QE as a function of the time at vacuum level of 4×10^{-6} Pa. The error bars indicate the maximum errors.

306 Figure 1
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Figure 2

Figure 3

Figure 4

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Figure 8

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Figure 9

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